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Spatial and dietary sources of elevated mercury exposure in white-tailed eagle nestlings in an Arctic freshwater environment☆

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ABSTRACT

Human-induced mercury (Hg) contamination is of global concern and its effects on wildlife remain of high concern, especially in environmental hotspots such as inland aquatic ecosystems. Mercury biomagnifies through the food web resulting in high exposure in apex predators, such as the white-tailed eagle (*Haliaeetus albicilla*), making them excellent sentinel species for environmental Hg contamination. An expanding population of white-tailed eagles is inhabiting a sparsely populated inland area in Lapland, northern Finland, mainly around two large reservoirs flooded 50 years ago. As previous preliminary work revealed elevated Hg levels in this population, we measured Hg exposure along with dietary proxies (δ^{13}C and δ^{15}N) in body feathers collected from white-tailed eagle nestlings in this area between 2007 and 2018. Mercury concentrations were investigated in relation to territory characteristics, proximity to the reservoirs and dietary ecology as potential driving factors of Hg contamination. Mercury concentrations in the nestlings (4.97–31.02 μg g^{-1} dw) were elevated, compared to earlier reported values in nestlings from the Finnish Baltic coast, and exceeded normal background levels (≤5.00 μg g^{-1}) while remaining below the tentative threshold of elevated risk for Hg exposure mediated health effect (>40.00 μg g^{-1}). The main drivers of Hg contamination were trophic position (proxied by δ^{15}N), the dietary proportion of the predatory fish pike (*Esox lucius*), and the vicinity to the Porttipahta reservoir. We also identified a potential evolutionary trap, as increased intake of the preferred prey, pike, increases exposure. All in all, we present results for poorly understood freshwater lake environments and show that more efforts should be dedicated to further unravel potentially complex pathways of Hg exposure to wildlife.

1. Introduction

Mercury (Hg) is a natural element that has been introduced into the environment in excessive amounts by anthropogenic activities at least since the 16th century. Today's environmental burden originates mainly from coal combustion and small-scale gold mining and have increased exponentially in the environment as well as in wildlife since the Industrial Revolution (Chen and Driscoll, 2018; United Nations Environment Programme, 2019). Anthropogenic Hg emissions have been distributed ubiquitously, even reaching remote ecosystems such as the Arctic (AMAP/UN Environment, 2019). Mercury is easily methylated in the aquatic environment (Jernelöv and Martin, 1975; Paranjape and Hall, 2017; Poulain and Barkay, 2013) and the resulting Hg species, methyl Hg (MeHg), is prone to bioaccumulation in biota followed by biomagnification through the food web (Atwell et al., 2011; Lavoie et al., 2013; Lehnherr, 2014). The highest concentrations are found in aquatic biota, and especially in top predators, integrating the contamination of their food web (AMAP/UN Environment, 2019; Scheuhammer et al., 2007). Sublethal adverse effects of Hg accumulation in animals have been detected in numerous studies and include reduction in reproductive success, neurological problems, failure in immune responses and avoidance of energy-demanding activities (Dietz et al., 2019; Evers, 2017; Whitney and Cristol, 2018).

The driving factors of Hg exposure and accumulation have been

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extensively studied in wildlife. Inter- but also intra-specific differences in Hg burden depend on diet, geographic location, large- and small-scale habitat characteristics and point sources of contamination (Lodenius and Solonen, 2013). The main diet factors affecting Hg burdens include trophic position (Lavoie et al., 2013; Lehnherr, 2014; Sebastiano et al., 2017) and the presence of fish species (Ackerman et al., 2016; Da Silva et al., 2005) or other aquatic prey in the diet (Barnes and Gerstenberger, 2015; Lodenius and Solonen, 2013). Large-scale habitat differences in Hg-burden occur between aquatic and terrestrial habitats (Ackerman et al., 2016) and freshwater and coastal habitats (Scheuhammer et al., 2007; Swanson et al., 2011). For terrestrial species, the Hg burden has been shown to increase in the proximity of watersheds (Barnes and Gerstenberger, 2015; Howie et al., 2018). Locally, also point sources such as coal-burning power plants and past military activity can result in increased Hg burdens (Anthony et al., 2007; Badry et al., 2019; Palma et al., 2005). The role of diet in Hg contamination is commonly studied using stable isotope ratios of carbon ($\delta^{13}C$) and nitrogen ($\delta^{15}N$). $\delta^{13}C$ is used to evaluate the origin of the prey, while $\delta^{15}N$ is an indicator for the trophic position of the prey (Boecklen et al., 2011; Peterson and Fry, 1987).

Positioned at the top of the food web, birds of prey are particularly prone to Hg accumulation and hence excellent sentinels for the contamination state of their environment (Helander et al., 2008). The white-tailed eagle (*Haliaeetus albicilla*, hereafter WTE) is a large raptor with a Palearctic distribution that mainly resides along watersheds. The Hg concentrations in WTEs in Northern Europe have historically been low (Berg et al., 1966; Dietz et al., 2006; Sun et al., 2019). They were however clearly elevated in the mid 20th century (mean 47.5 μg g^{-1}, Sun et al., 2019) around the highly contaminated Baltic Sea (Leipe et al., 2013). Since then, the Hg levels in the area have fallen to 14.1 μg g^{-1} and continues to be at background levels at the Atlantic coast in Greenland, Norway and Poland (Kalisinska, 2014; Kitowski et al., 2017; Sun et al., 2019). A recent preliminary study from Finland reported low Hg concentrations in feathers of WTE nestlings at multiple locations in the Finnish Baltic coast (mean < 5 μg g^{-1}), while the few nestlings from inland northern Finland, Lapland, had elevated Hg concentrations (mean 9.67 μg g^{-1}, Johansson, 2019). These first measures of Hg concentrations in WTE nestlings hatched in Lapland were relatively high compared to other observations on nestlings in the Nordic environment and therefore warranted further investigation as the cause for the elevated concentrations were unknown. The WTE population in Lapland also provides an excellent opportunity to examine different drivers of Hg contamination as both the local landscape and the diet composition of the nesting pairs is diverse (Ekblad et al., 2020). Moreover, the core area of this population is situated around two large artificial water reservoirs. Mercury levels are known to be elevated in water reservoirs, especially in newly flooded ones where the oxygen-free conditions in the submerged soils brace methylation by microbes, but the levels can still be elevated decades later (Friedl and Wüest, 2002; Hsu-Kim et al., 2018; United Nations Environment Programme, 2019; Verta et al., 1986). Mercury contamination in fish and other aquatic organisms in water reservoirs is well studied (e.g. Li et al., 2015; Surma-Aho et al., 1986; Wang and Zhang, 2013; Willacker et al., 2016), while little is known about the transfer to terrestrial species foraging in and inhabiting the surroundings of artificial reservoirs, even though terrestrial ecosystems mainly receive their MeHg through biotransporters from the aquatic food webs (Schiesari et al., 2018).

Mercury in animals has been measured from a variety of tissues. The advantage of feathers is that the sampling can be done with minimal invasiveness for the nestlings (Eulaers et al., 2011a; Eulaers et al., 2011b; Lodenius and Solonen, 2013). Furthermore, Hg deposited in growing feathers upon nestling growth integrates environmental levels through consumption of prey gathered by the parents in the nesting territory in contrast to adults, whose feathers reflects the Hg exposure of the moulting area (Fuchsman et al., 2017). As such, sampling and analysing an entire feather of a nestling close to fledging results in a chemically stable archive of the Hg exposure and its dietary pathway during the majority of the nestling stage (Eulaers et al., 2011a; Eulaers et al., 2011b). In addition, variation among different body feathers has been reported to be rather small in nestlings (Carravieri et al., 2014), further promoting the promising use of nestlings and their body feathers to study environmental Hg contamination and pathways. Finally, Hg in feathers is mostly methylated, allowing to assume that the total Hg concentration in the feather is MeHg (Bond and Diamond, 2009).

The aims of the present study were 1) to examine the magnitude of feather Hg concentrations in the Finnish WTE Lapland population as preliminary data indicated elevated levels (Johansson, 2019); and 2) to quantify the relative importance of potential driving factors such as abiotic point sources (i.e. the artificial reservoirs), territory characteristics, long-range transport and dietary plasticity. To our knowledge, this is the first study to investigate feather Hg burdens at the individual level in a WTE population with regards to nestling diet and habitat characteristics of the breeding territory. On the basis of available literature, we hypothesized that the Hg concentrations are higher in WTE nestlings eating more fish, feeding on a higher trophic level, originating from territories with more marshland and water where the fish consumption is higher, and in territories situated close to the reservoirs.

2. Material and methods

2.1. Study area, study species and sampling

The study area is situated in Northernmost Finland, in the scarcely inhabited province Lapland (Fig. 1). It is characterized by low forests, peat bogs and inland waters. The dominating tree species is scots pine (*Pinus sylvestris,* 77% of the forested area) followed by Norway spruce (*Picea abies,* 16%) (Natural Resources Institute Finland, 2020). The largest water bodies in the area are the two reservoirs of Lokka and Porttipahta, flooded in 1967 and 1971, respectively (Hellsten et al., 1993), situated approximately 20 km from each other.

The white-tailed eagle (*Haliaeetus albicilla*) is a large diurnal territorial raptor that defends a nesting territory, on which it has one or multiple alternative nests it chooses between in different years (Cramp, 1980). It hunts in the vicinity of the nest, mainly within a 10 km radius (Ekblad et al., 2020; Krone and Treu, 2018). In Lapland, prey remnant investigations have shown its diet to consist of 65% fish, mainly northern pike (*Esox lucius*), 28% birds that are mainly waterfowl, grouse, and cranes, and of a minority of mammals, including reindeer calves (*Rangifer tarandus*) (Ekblad et al., 2020; Sulkava et al., 1997).

The territory of a WTE pair was determined as an area of a 10 km radius around the recorded GPS coordinates of their nest, or nest site, if there were multiple nests in the territory. The proportions of different landscape types within this area were calculated using landcover data from the open source CORINE 2012 with a resolution of 20 × 20 m (Finnish Environment Institute, 2019). The diet information of the WTE pairs is based on identification of prey remains collected at the nests. For more detailed descriptions about the territories and prey data we refer to Ekblad et al. (2020). Of the available data, we considered only territories where at least five prey items had been recorded.

The WTEs that nest in Finland are surveyed annually since 1973 according to a national monitoring program run by volunteers (Högmander et al., 2020). In Lapland, the survey of WTEs is governmental and conducted by the state forest administration Metsähallitus (Nature conservation decree). As Lapland is a large and very sparsely inhabited area with only a few roads, the breeding status is first checked by helicopter, after which the nests with nestlings are visited from the ground when the nestlings are approximately three to eight (mean six) weeks old. The nestlings are ringed and measured, and 3–5 body feathers are gently plucked from the back of each nestling. These body feathers are not necessarily fully-grown at this age, though have been growing for several weeks. See Table 1 for an overview of the nestlings in the present study. The feathers are transported in paper envelopes at

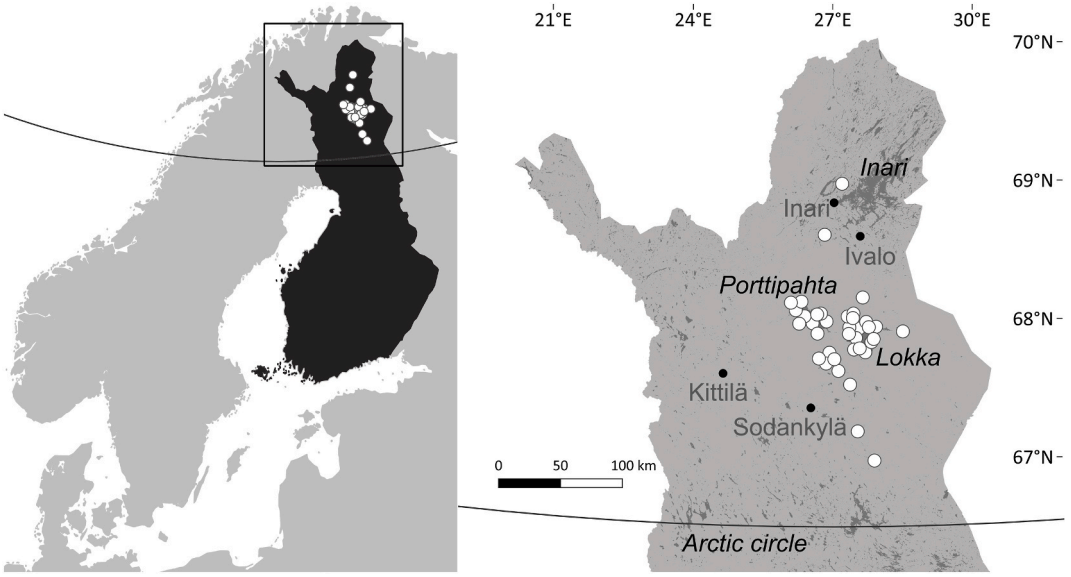

Fig. 1. Study area in Lapland, northern Finland, and locations of the sampled white-tailed eagle nests (white dots) within the area. The artificial water reservoirs Porttipahta and Lokka, around which the majority of the WTEs in the area nests, are shown on the map along with the natural lake Inari (*in italics)*.

Table 1
Temporal distribution of the white-tailed eagle nestlings in the present study. The values regard the total number of nestlings measured for Hg (μg g^{-1} dw) and isotopic ratios (‰). In parentheses are the number of nestlings that originate from territories for which there are available diet data (prey items tot ≥5).

Year	*n* territories	*n* nestlings	Mean Hg (±sd)	Mean δ^{15}N (±sd)	Mean δ^{13}C (±sd)
2007	4 (4)	6 (6)	13.97 ± 2.32	11.57 ± 0.37	−27.85 ± 0.37
2008	1 (1)	2 (2)	6.64 ± 1.02	8.82 ± 0.13	−23.68 ± 0.17
2010	2 (2)	2 (2)	8.58 ± 1.85	10.90 ± 0.10	−26.13 ± 3.56
2011	2 (2)	2 (2)	13.60 ± 9.19	10.62 ± 0.71	−28.32 ± 0.90
2012	1 (1)	1 (1)	14.96	11.70	−27.19
2014	19 (15)	27 (20)	12.95 ± 5.39	11.91 ± 1.19	−26.58 ± 1.73
2015	7 (5)	8 (5)	15.42 ± 10.03	11.61 ± 0.83	−26.07 ± 1.05
2016	12 (10)	13 (11)	9.62 ± 3.13	11.81 ± 1.46	−26.48 ± 1.64
2017	14 (9)	17 (11)	13.57 ± 3.91	12.35 ± 1.20	−27.23 ± 1.24
2018	7 (6)	7 (5)	10.66 ± 5.49	11.35 ± 1.61	−26.06 ± 1.98
Total	37 (24)	85 (65)	12.47 ± 5.43	11.76 ± 1.28	−26.66 ± 1.63

ambient temperature and humidity and moved to −24 °C for long-term storage.

2.2. Sample preparation

Of the collected feathers, only one of each nestling was made available for the present study and therefore asked for an analytical plan that would deliver both reliable Hg concentrations and stable carbon and nitrogen isotope values. From each body feather the calamus and any present down was removed to not bias Hg concentrations by dissimilar growth periods (Bontempo et al., 2014; Bortolotti, 2010) or potential in ovo exposure (Ackerman et al., 2008; Monclús et al., 2018), respectively. For four young chicks only downy feathers were present, in which case these were used. Each body feather was consequently thoroughly washed using distilled water until visual confirmation that the washing solution (refreshed when deemed needed) was not further contaminated upon agitation of the vanes. After washing, the feathers were covered with analytical-grade laboratory paper and left to dry overnight at ambient room temperature in a controlled laboratory environment. Each feather was then cut into 1 mm pieces using stainless steel scissors.

Even though washing with distilled water is not conventional for stable isotope analysis, this was a pragmatic and informed choice due to having to carry out both Hg and stable isotopes on the same limited feather material, as much as it was a decision that ad hoc a more aggressive washing method to remove lipids was not required. Firstly, body feathers of WTE nestlings are not considered to be covered with high lipid preen oil due to the low preening intensity of this life stage, the physical distance between the sampled dorsal body feathers and the preen gland, and finally an overall much lower preening intensity in birds of prey than in seabirds. The absence or very low presence of lipids on WTE nestling body feathers, as well as the lack of the need to normalize among individuals, was demonstrated by earlier results as well as ad hoc by low and stable C:N ratios of 3.14 ± 0.05 (mean ± SD). Secondly, while the presence of lipids has indeed been shown to bias δ^{13}C values (Boecklen et al., 2011; DeNiro and Epstein, 1978; Post et al., 2007), to the best of our knowledge, this is not the case for δ^{15}N values. Moreover, the presence of preen oil on the feather surface is not considered problematic, rather beneficial, for feather Hg to reflect internal body burdens (see Jaspers et al., 2019 and references therein). Washing with (a mixture of) organic solvents, even when followed by rinsing with distilled water, also impacts δ^{15}N values (e.g. Paritte and Kelly, 2009; Valladares et al., 2010). As in the present study a shift in δ^{15}N values was considered more detrimental (the biomagnification aspect using δ^{15}N as trophic proxy) than a shift in δ^{13}C values (to discriminate lacustrine from terrestrial sources) we decided to not use the more conventional procedure of using (a mixture of) organic solvents. However, as a result of our preparation method deviating from conventional methods using (a mixture of) organic solvents some caution may be warranted when comparing absolute stable isotope

values.

2.3. Mercury analysis

The analysis for total Hg content in the nestling feathers was performed at the accredited trace element laboratory of the Department of Bioscience, Aarhus University (Denmark). A subsample of the homogenized feather (see section 2.2), ranging from 1.5 to 10.9 mg, was analysed for total Hg content (μg g^{-1}) on a dry weight (dw) basis using a Milestone DMA-80 Direct Hg Analyzer (Sorisole, Italy) following the U. S. EPA Method 7473 (EPA, 1998). The instrumental analytical quality control was verified by concurrent analysis of procedural blanks, duplicates, aqueous standards (10 ng and 100 ng Hg, Sigma-Aldrich, Switzerland), and Certified Reference Material (CRM; DORM-4, National Research Council, Ottawa, Canada). All samples and CRMs were corrected for the average blank amount of Hg as well as for the recovery of aqueous standards. Measurement of CRMs revealed satisfactory accuracy (recovery of 105 ± 5%; $n = 17$) in line with the certified value (0.410 ± 0.055 μg g^{-1} dw). Duplicate analysis of the same feather showed good precision with RSD ≤7.79% ($n = 5$). All body feather concentrations are expressed in μg g^{-1} dw.

2.4. Stable isotope analysis

The analysis for stable carbon (δ^{13}C and ^{12}C) and nitrogen isotopes (^{15}N and ^{14}N) was carried out at the Stable Isotope Lab of the University of Koblenz-Landau (Germany). Ratios of carbon (^{13}C:^{12}C) and nitrogen (^{15}N:^{14}N) in a subsample (mean ± *SD*: 1.48 ± 0.06 mg) of each homogenized feather (see above) was determined using a Flash 2000 HT elemental analyzer coupled via a ConFlo IV interface to a Delta V Advantage isotope ratio mass spectrometer (all Thermo Fisher Scientific, Bremen, Germany). The stable isotope values for carbon and nitrogen are conventionally expressed as δ values (‰) relative to their respective international measurement standards Vienna Pee Dee Belemnite and atmospheric N_2, respectively. Internal reference material (casein) was measured concurrently in duplicate every ten samples, revealing an imprecision (±*SD*) ≤ 0.06‰ for both δ^{13}C and δ^{15}N. Analysis of duplicate feather samples showed good precision for both stable carbon (RSD ≤1.34%) and nitrogen isotopes (RSD ≤3.56%).

2.5. Model selection

Based on prior information about drivers of Hg contamination, we formulated a set of candidate models. Prior to constructing the models, we examined whether there were any time trends or a correlation between the Hg concentration and the age of the nestlings. Since there was no time trend in Hg concentrations and they were not explained by wing length (as determined by a linear mixed model with Hg concentration as response variable and year and wing length (a surrogate of age) as predictors ($n = 77$); estimate year = 3.33 ± 3.42, $t_{12} = 0.97$, p = 0.35, estimate wing = −4.73 ± 3.67, $t_{67} = 1.29$, p = 0.20), these variables were not considered in the model selection procedures. However, there was variation between the years, potentially due to sampling at different types of territories in some years from just a few nestlings. Year was thus included as a categorical random effect. The candidate models representing different hypotheses included 1) variation in the dietary habits (proxied by δ^{13}C and δ^{15}N, or proportion of pike and fish), 2) geographic location, 3) variation in habitat use (proportion of lakes and marshlands in the territory), 4) proximity to the nearest reservoir and 5) a full model with all the factors. We give the reasoning behind these hypotheses and the corresponding models below.

1. **Di**etary ecology model. Mercury accumulates in food webs and especially in the aquatic element (Lavoie et al., 2013). We therefore used diet variables testing for these aspects.
   - δ15N and δ13C: these respective stable isotope values are established proxies for the trophic position or food web origin (Peterson and Fry, 1987). We expected WTEs feeding at a higher trophic position to exhibit a higher Hg exposure, and WTEs with values indicative for increased aquatic feeding to have higher Hg exposure.
   - **Proportion of fish, especially pike.** In birds, a fish-rich diet is associated with higher Hg exposure (Ackerman et al., 2016). Especially pike, which is a long-lived top-predator in the aquatic environment and the major prey of WTEs in the study area, tends to bioaccumulate Hg (Pierce, 2012).
2. Long-range transport model.
   - **Latitude.** Mercury exposure seems to be elevated in higher latitudes (Lavoie et al., 2013), which could result in higher Hg contents of WTEs originating from further north. Contradictory, Hg concentrations in freshwater fish in Fennoscandia tend to decrease towards higher latitudes (Ahonen et al., 2018; Braaten et al., 2017). The latitudinal range of territories is 224 km and the longitudinal 90 km. The longitudinal gradient is highly influenced by the Lokka reservoir being situated east of Porttipahta, which is why we did not include longitude in the model but rather examine the difference between the reservoirs (see the Point source model below).
3. Territory habitat model.
   - **Proportion of lakes and marshlands in the territory.** The proportion of pike and other fish in the diet of WTEs increases with increasing proportion marsh or lakes in the territory (Ekblad et al., 2020). We expected WTE nestlings from territories with larger proportions of marsh and lakes to have higher Hg exposure.
4. Point source model.
   - **Reservoir proximity.** Man-made reservoirs have elevated Hg levels up to decades after being flooded (Verta et al., 1986). We defined a three-level variable with levels 'closer to Lokka', 'closer to Porttipahta' and 'Elsewhere'. Territories situated >15 km from the reservoirs were assigned to the category Elsewhere. The distance between the reservoirs is approximately 20 km at the closest ends. The median distance to Porttipahta is 0.98 km for territories with diet information ($n = 10$) and 0.67 km for territories without diet information ($n = 7$), while the median distance to Lokka is 0.58 km for both ($n = 17$ and 11, respectively). Territories assigned to be Elsewhere are situated 16–100 km away from the reservoirs, with medians 26 km ($n = 10$) and 23.7 km ($n = 6$).
5. Full model.
   - In the full model we included all the factors from the models above that could be used without violating collinearity (section 2.5).

Data on Hg and latitude, habitat characteristics, δ^{13}C and δ^{15}N was available for all 85 studied nestlings, while diet information (where n tot ≥5) was available for 65 nestlings. Model selection needs to be done with exactly the same data and therefore the data on the 65 nestlings were used in all the models to be able to compare them.

2.6. Statistical analysis

All statistical tests were performed using R version 3.6.2 (R Core Team, 2020). We fitted linear mixed models with the package *lmerTest* (Kuznetsova et al., 2017) with the Hg concentration as response variable and the model variables described above as predictors. As some of the nestlings originated from the same territories and the data was collected over a time span of 12 years, territory ID and year were added as random effects in all models. The models were explored for normality, heteroscedasticity and influential outliers by residual plots, Shapiro-Wilk's test, Barlett's test and Cook's distance. Collinearity of the explanatory variables was tested with the Variance Inflation Factor (VIF) with a threshold of 3.00 (Zuur et al., 2010). All continuous variables were scaled to a value between 0 and 1 before fitting the models (((variable -

min (variable))/(max (variable)-min (variable))) or ((percentage fish, pike, or habitat in territory)/100)). The goodness of the models was compared by Akaike's Information Criterion corrected for small sample sizes (AICc; Burnham and Anderson, 2002). Post-hoc comparisons of the categorical variable were performed with Tukey's test with the package lsmeans (Lenth, 2016).

The diet predictors $\delta^{15}N$ and $\delta^{13}C$ were negatively correlated with each other ($r = -0.72$), indicating that aquatic prey mainly originated from a higher trophic level than terrestrial prey, while the proportions of fish and pike were positively correlated ($r = 0.67$). The VIFs were nevertheless under the threshold of 3.00 (Zuur et al., 2010) and the variables were hence simultaneously used as explanatory variables in the diet model. In the full model, $\delta^{13}C$ was excluded to avoid collinearity issues (VIF = 4.14). Hence, the predictors in the full model were $\delta^{15}N$, proportion pike, proportion fish, latitude, proportion water, proportion marsh and reservoir proximity (Online Resource 1).

Confidence intervals (95%) for the graphs were obtained by bootstrapping with the bootMer function from the lme 4-package (Bates et al., 2015), where new data from the fitted model was simulated 200 times and the simulations that fell within ±47.5% from the mean was used.

To verify that the achieved accuracy of the analysis, performed on one feather only, does not result in inaccurate Hg concentrations due to intra-individual variations, we performed some additional tests. The obtained Hg concentrations were randomly changed 0 - ± 20% to simulate the measurement error of median 10% proposed by Peterson et al. (2019). This was repeated 100 times, and the best model was re-run 100 times with these new values.

3. Results

3.1. Feather Hg concentration

The geometric mean Hg concentration measured in the nestlings was 11.48 μg g^{-1} (4.97 μg g^{-1} – 31.02 μg g^{-1} dw). One nestling originating from a nest close to Porttipahta had considerably lower Hg concentrations (5.39 μg g^{-1} dw) than other nestlings from previous and subsequent years in this territory (20.10, 22.77 and 21.35 μg g^{-1} dw). This observation was detected as highly influential in the models by the Cook's distance test and led to heteroscedasticity in the predictor 'proximity to reservoir', with level 'Porttipahta' having more spread. We did not however exclude it, as it had only a minor impact on the results and in such cases the recommendation is to keep it (Frost, 2019). The two nestlings with the highest Hg concentrations (>30 μg g^{-1}, the third highest being 27.11 μg g^{-1} dw) were siblings originating from the same nest in the same year. There was no diet information for these nestlings, so they were automatically excluded from the further analyses. The nest was located 12.5 km upstream of Lokka, which did not fit in to the general pattern described below.

3.2. Driving factors of Hg contamination

Of the *a priori* defined models, the full model gained the most support, but the diet model was virtually as good (ΔAICc = 0.19; Table 2). The other models (point source, long range, or habitat) were less supported (Table 2). Examination of the full model indicated that it included four variables that had no obvious effect on the observed variation in Hg concentrations (proportion of lakes in the territory: p = 0.29; proportion of marsh: p = 0.62, latitude: p = 0.60 and proportion of fish in the diet: p = 0.64). We therefore removed these variables, gaining an *a posteriori* model (6) that was considerably better than the full model (delta AICc = −8.71). This model showed that Hg increased with $\delta^{15}N$ and the proportion of pike in the diet and was higher in the territories close to Porttipahta than in territories close to Lokka or far from the reservoirs (Fig. 2, Table 3). Since Restricted Maximum Likelihood is considered to give better estimates for the random effects than Maximum Likelihood, this model was run with REML for final inference of the estimates (Table 3).

Table 2
Results of model selection for candidate models explaining Hg concentrations in white-tailed eagle nestlings from Finnish Lapland in 2007–2018. In the explanatory variables 'prop.' stands for proportion of diet items or habitat in the nesting territory. Reservoir is a three-level variable that indicates whether the territory is closer to Porttipahta or Lokka, or is situated Elsewhere. $n = 65$ individuals from 24 territories. Territory ID and Year are included as random effects. The models are sorted by AICc, the most parsimonious model having the lowest value, and the difference is indicated in the table by ΔAICc. *w* shows the Akaike's weight (Burnham and Anderson, 2002).

Nr.	Model	Variables	ΔAICc	*w*	R^2
6.	*A posteriori*	$\delta^{15}N$ + proportion pike + Closest reservoir	0	0.976	0.57
5.	Full model	$\delta^{15}N$ + prop pike + prop fish + Closest reservoir + prop lake + prop marsh + latitude	8.71	0.013	0.58
1.	Diet	$\delta^{15}N$ + $\delta^{13}C$ + prop pike + prop fish	8.90	0.011	0.48
4.	Point source	Closest reservoir	21.58	0	0.35
2.	Long-range	Latitude	35.37	0	0
3.	Habitat	Prop lake + prop marsh	37.71	0	0

The results of the additional tests, in which the Hg values randomly were changed 0 - ± 20% to simulate possible measurement errors, were consistent with these results and validated our conclusions based on only one feather per nestling (Online Resource 2).

4. Discussion

4.1. Mercury burden of the WTE nestlings

The Hg concentrations in WTE nestlings from Finnish Lapland were three times higher than in WTEs originating from the Baltic coast of Finland (Johansson, 2019). Only one nestling showed exposure lower than 5 μg g^{-1} dw, generally considered the natural biogeochemical background level reflected in raptor feathers (Scheuhammer, 1991). No nestlings, however, showed concentrations exceeding 40 μg g^{-1} dw, which has been proposed to be the threshold level for adverse effects in this species (Sun et al., 2019). Generally, the main drivers of Hg contamination for the nestlings were found to be dietary habits as well as proximity to the Porttipahta reservoir acting as an environmental point source.

4.2. Factors influencing Hg exposure

As hypothesized, due to the biomagnification of MeHg (Atwell et al., 2011; Peterson and Fry, 1987), nestlings fed with higher trophic prey (as proxied by $\delta^{15}N$) exhibited higher Hg concentrations in their feathers. This was also by far the most important factor (Table 3). The other diet factor affecting Hg concentration was the proportion of pike. Although the Hg burden in apex predators almost exclusively has a dietary origin (Shore et al., 2011) and the Hg concentrations in freshwater fish in Fennoscandia tend to decrease towards higher latitudes (Ahonen et al., 2018; Braaten et al., 2017), some factors are creating a potential hotspot for Hg accumulation in WTEs in Lapland. In polar regions and especially in freshwater ecosystems, Hg biomagnification per trophic level is higher than in lower latitudes (Lavoie et al., 2013), which accentuates the significance of trophic level in this system. Slow-growing fish accumulate higher Hg concentrations than fast growing (Simoneau et al., 2005), which can explain the higher bioaccumulation in polar regions (Lavoie et al., 2013). Furthermore, Hg concentrations are higher in larger fish specimens, especially pike (Ahonen et al., 2018; Łuczyńska, 2005) and WTEs select larger fish as prey when available

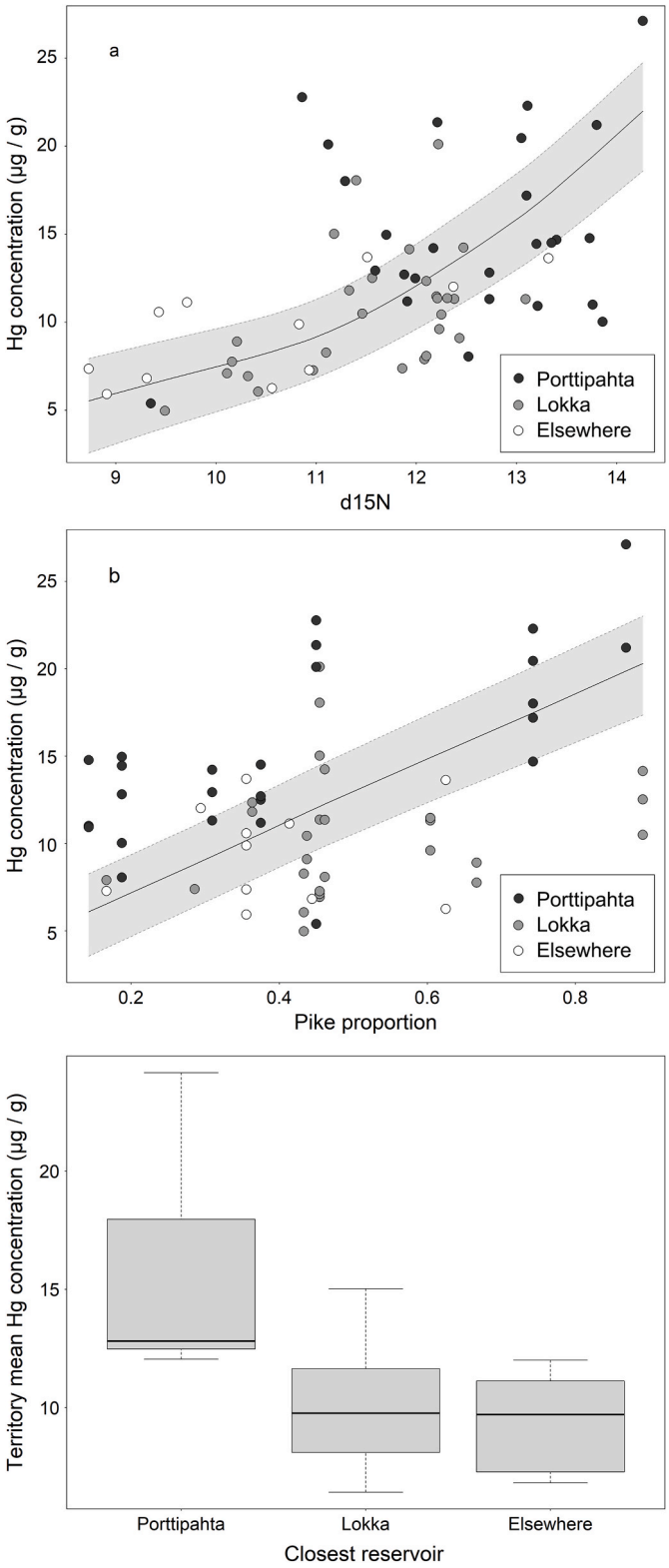

Fig. 2. Mercury concentration in body feathers of white-tailed eagle nestlings ($n = 65$) in Finnish Lapland in 2007–2018 in relation to a) $\delta^{15}N$ in the feathers of the nestlings and b) proportion of pike in the diet of the territory. The shaded area shows 95% confidence intervals obtained by bootstrapping. The dots are actual observations and are colored according to which reservoir (Lokka or Porttipahta) the nest is closer to (or far from both, i.e. Elsewhere). c) Mean territorial ($n = 36$) Hg concentration in nestlings in relation to which reservoir the nest is closer to. The territory near Lokka with extreme concentrations is excluded from the figure.

Table 3
The linear mixed model best explaining variation in Hg concentration of white-tailed eagle nestlings in Finnish Lapland 2007–2018. Year and Territory ID were included as random effects and $n = 65$ nestlings from 24 territories.

Effect	Estimate	DF	*F*	*P*
$\delta^{15}N$	1.89 ± 0.40	1, 59.3	22.43	>0.0001
proportion pike	7.74 ± 2.83	1, 21.5	7.48	0.012
Closest reservoir		2, 24.9	6.10	0.007
Lokka	0			
Elsewhere	2.40 ± 1.55			
Porttipahta	4.62 ± 1.34			

(Nadjafzadeh et al., 2016). The combination of these factors results in WTE individuals feeding on higher trophic levels and higher proportion of pike accumulating disproportionately high Hg concentrations. No impact of habitat characteristics or latitude were observed.

These results highlight the importance of considering multiple factors and paying attention to local circumstances when examining Hg contamination, as very different patterns have emerged in different studies. For example, temporal trends are mainly detected in studies spanning several decades with substantial reductions or increases in environmental contamination (Dietz et al., 2009; Sun et al., 2019). Interspecific differences are predominantly, but not always, better explained by $\delta^{15}N$ than $\delta^{13}C$ in (Anderson et al., 2009; Einoder et al., 2018; Hebert, 2019). In contrast, for Arctic birds, Anthony et al. (2007) showed that neither of the isotopes were important but instead attributed Hg concentrations to long-distance air deposition and point sources. Intraspecific differences for peregrine falcons in Portugal were driven by the distance to water and proportion of aquatic prey (Barnes and Gerstenberger, 2015). A study comparing multiple contamination pathways for Bonelli's eagles in Portugal (Badry et al., 2019) identified similar factors important as the present study; mainly $\delta^{15}N$ and the distance to power plants along with artificial structures, while $\delta^{13}C$ and the habitat factors were identified as non-important. Regarding Hg contamination in terrestrial species living close to reservoirs, Ospreys hatching near a reservoir in Canada had five times higher Hg concentrations than ospreys originating from territories further away (Desgranges et al., 1998), and the proximity of a reservoir had a small elevating impact on otter fur Hg concentrations (Crowley and Hodder, 2019; Spencer et al., 2011).

4.3. Fish as primary Hg source might be an evolutionary trap

Fish have been shown to be the preferred prey of WTEs in Germany (Nadjafzadeh et al., 2013) and presumably in Lapland (Ekblad et al., 2020). In the light of this, it is of some concern that the elevated Hg concentrations are tied to fish consumption. This might actually be an evolutionary trap that takes place when an organism makes a choice that previously was adaptive but no longer is due to recent anthropogenic alterations (Schlaepfer et al., 2002). Evolutionary traps have been identified in terms of disadvantageous foraging behaviour, navigation, oviposition and mate selection (Robertson et al., 2013). To our knowledge, this concept has hardly been discussed for contaminated prey. Herring et al. (2020) proposed an ecological trap for Golden eagles in the US nesting close to shooting fields where they fed on Pb contaminated prey. A switch to a more contaminated food source has been found for seabirds that chose trawler discards over their normal diet, even though the discards originate from deeper waters and have significantly higher Hg burdens (Arcos et al., 2002). For the WTEs in Lapland the case is somewhat different, as their presumably preferred food (fish) appears to be relatively higher contaminated compared to alternative prey (birds and mammals). The WTEs in the Baltic would have experienced a corresponding situation in the 1960s and 1970s, when the Hg contamination in the Baltic food web was considerable (Helander et al., 1982; Sun et al., 2019). However, in the Kvarken area in the middle of the Gulf of Bothnia, fish-eating waterfowl were the major source of Hg while the

local pike had low Hg concentrations (Koivusaari et al., 1975). It is important to note that not all requirements for an evolutionary trap are yet met in this study, as there are no measurements on the fitness outcome of the eagles regarding diet differences (Robertson et al., 2013). Our results nevertheless warrant further investigation of this potential issue.

4.4. Freshwater reservoirs as a source for legacy Hg

In Lapland, the background Hg levels (as assessed from mosses and the atmospheric deposition) are generally low (AMAP/UN Environment, 2019; Poikolainen and Rautio, 2015). Atmospheric Hg deposition trends are however often not consistent with accumulation in local biota. This phenomenon is particularly evident for freshwater reservoirs, offering conditions that drive conversion of Hg to MeHg and eventually the availability in the food web (AMAP/UN Environment, 2019). The persistency of Hg in freshwater reservoirs varies considerably but is commonly thought to be in the time span of 5–25 years (Lodenius et al., 1983). However, a more recent study from North America showed that Hg concentrations in fish were slightly elevated in some reservoirs compared to natural lakes still a century after the impoundment (Willacker et al., 2016), depending on the hydrology and timing of water storage peak in the reservoir (e.g. Mir et al., 2020; Ullrich et al., 2001; Willacker et al., 2016). Measurements from the half-century-old Lokka and Porttipahta reservoirs indicate that the mean Hg concentrations in pike were close to those in natural lakes already in the 1990's (Lokka 0.29 μg g^{-1}, Porttipahta 0.41 μg g^{-1} (Porvari, 1998)). They continued to decrease until around 2010, after which the concentrations have stabilized (Pöyry Finland Oy, 2017). The Hg concentrations have consistently been higher in Porttipahta than in Lokka (Huttula, 2006; Mehtälä, 2014; Porvari, 1998) even though the latest report from 2016 (Pöyry Finland Oy, 2017) did not detect this, probably due to the insufficient sampling of large fish and labelling errors.

The elevated Hg-concentrations in the WTE nestlings from Lapland investigated in the present study did not show any decreasing trend during the time period 2007–2018. Hence, the source of the Hg is still present in the ecosystem and our results strongly indicate that WTEs get the highest concentrations from pike in the Porttipahta reservoir. Large pike and other long-lived predatory fish might still possess substantial Hg burdens, which is mirrored into the food web. The legacy Hg carried by the reservoir is a hotspot for Hg contamination in the area, currently overseen by the compulsory mainstream investigations without requirement for different size-classes of fish. However, there are also other unknown sources of Hg contamination in the system than the ones we were able to identify. The two siblings originating from the surroundings of the lake Kopsusjärvi, from a nest situated 12.5 km upstream from Lokka, had the highest Hg concentrations of all measured chicks (30–31 μg g^{-1}) while their isotope values were mediocre for both $\delta^{13}C$ and $\delta^{15}N$. The nest is in a nature conservation area with little apparent human activity in the vicinity, and it is thus unclear what the source of Hg could be. The second highest concentrations close to Lokka (15–20 μg g^{-1}) were found in a territory located downstream of Lake Kopsusjärvi, indicating that the Hg levels in this explicit area indeed are elevated. Mercury measurements along with Hg isotope analysis (Blum et al., 2014) of wildlife originating from watersheds in that area should be made to trace the possible sources.

4.5. Health risk associated with Hg exposure

As the feather Hg concentrations of the examined WTE nestlings were below 40 μg g^{-1} no immediate health risk is assumed. However, with some detected concentrations as high as 31 μg g^{-1} a rise in environmental Hg concentration in the area could cause some WTE individuals to accumulate potentially harmful concentrations. It has however also been shown that the related bald eagles (*Haliaeetus leucocephalus*) are relatively efficient in demethylating Hg using selenium (Se) (Scheuhammer et al., 2008), a trait that many piscivorous apex predators coping with high dietary Hg levels appears to have, making them less vulnerable to Hg toxicity (Eagles-Smith et al., 2009; Fuchsman et al., 2017; Scheuhammer et al., 2008). For this reason, we also recommend further research to jointly determine Hg and Se in both predator and prey species to properly evaluate a potential health risk.

5. Conclusions

The main cause of elevated Hg concentrations in WTE nestlings in Finnish Lapland is bioaccumulation from a diet of prey from high trophic levels; long-lived pike, originating from an artificial lake that carries a legacy of high Hg levels into the present day. Our results indicate that Hg levels are still elevated in the biota in the Porttipahta reservoir, which leads to higher burdens also in the WTEs outside the aquatic environment. In particular large pike are likely to contain high concentrations of Hg, which cause pike-eating WTE nestlings to bioaccumulate Hg concentrations close to proposed risk threshold levels. Our results highlight the promising potential of WTE nestlings to act as sentinel species for local habitat conditions and the importance of considering multiple drivers when dealing with Hg contamination. We also identified a potential evolutionary trap for the WTEs, whose preferred prey has been shown to be (large) fish. WTEs implicitly feeding on their preferred prey are the ones that accumulate the highest Hg concentrations. Mercury concentrations in some WTE individuals within the study were close to proposed health effect levels, which highlights the importance for future more in-depth monitoring of WTEs as well as their constituting food web components in the study area.

Credit author statement

Camilla Ekblad: Conceptualisation, Methodology, Formal analysis, Investigation, Data curation, Writing – original draft, Visualisation, Project administration, Funding acquisition, Igor Eulaers: Conceptualisation, Methodology, Validation, Resources, Writing – review & editing, Supervision, Project administration, Ralf Schulz: Resources, Writing – review & editing, Funding acquisition, Torsten Stjernberg: Resources, Data curation, Writing – review & editing, Jens Søndergaard: Resources, Writing – review & editing, Jochen Zubrod: Validation, Investigation, Writing – review & editing, Toni Laaksonen: Conceptualisation, Methodology, Writing – review & editing, Supervision, Project administration, Funding acquisition

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envpol.2021.117952.

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